

Spin-orbit coupling effect in (Ga,Mn)As films: anisotropic exchange interactions and magnetocrystalline anisotropy

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The magneto-crystalline anisotropy (MCA) of (Ga,Mn)As films has been studied on the basis of ab-initio electronic structure theory by performing magnetic torque calculations. An appreciable contribution to the in-plane uniaxial anisotropy can be attributed to an extended region adjacent to the surface. Calculations of the exchange tensor allow to ascribe a significant part to the MCA to the exchange anisotropy, caused either by a tetragonal distortion of the lattice or by the presence of the surface or interface.

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Diluted magnetic semiconductors (DMS) are a class of materials having attractive properties for spintronic applications (e.g. see review [1]). Many investigations in this field are focussed on the (Ga,Mn)As DMS system with 1 to 10% of Mn atoms which have promising features from a physical as well as technological point of view. The crucial role of valence states with respect to various magnetic properties of (Ga,Mn)As was discussed in the literature by many authors [1, 2]. First of all, the valence band holes are responsible for ferromagnetic (FM) order in the system mediating the exchange interaction between well localized Mn magnetic moments. Spin-orbit coupling of the states at the top of valence band, being close to the Fermi level, leads to a rather strong cubic magnetocrystalline anisotropy (MCA) in bulk (Ga,Mn)As and to an in-plane biaxial MCA in the (Ga,Mn)As film on top of a GaAs substrate [3]. In the latter case the spin-orbit coupling (SOC) makes the valence states close to E_F sensitive to lattice distortions and in that way responsible for the in-plane MCA due to compressive strains originating from the lattice mismatch between the (Ga,Mn)As film and GaAs substrate [4–14]. As soon as the spin polarization of the valence bands is rather small, the MCA in (Ga,Mn)As is discussed in terms of anisotropic exchange interactions of the Mn atoms [2–4]. The strength of the MCA depends on the hole concentration introduced by the Mn impurity atoms [3, 8, 15, 16] as well as on the variation of equilibrium lattice parameter of (Ga,Mn)As increasing with the increase of Mn content and resulting in a larger lattice mismatch with the GaAs substrate.

Numerous experimental results evidenced a transition from the bi-axial to the uni-axial in-plane anisotropy [8–15]. So far, however, there is no consensus in the literature concerning the origin of the in-plane uniaxial anisotropy. Although in some recent theoretical works the origin of the uniaxial in-plane anisotropy is attributed to a trigonal distortion caused by a uniaxial or shear strain within the film plane [10, 15, 17], this type of distortion was not observed experimentally. Stacking fault

defects in the (111) and (11 $\bar{1}$) planes have been found recently in experiment [18] which could be responsible for breaking the equivalence of the [110] and [1 $\bar{1}$ 0] directions in the (Ga,Mn)As films. However, there is so far no experimental evidence nor theoretical description showing that these stacking faults are responsible for the in-plane uniaxial anisotropy.

In order to obtain a more detailed understanding of the subtle electronic effects which determine the MCA properties of (Ga,Mn)As films, we performed ab-initio electronic structure calculations for tetragonally distorted (Ga,Mn)As bulk as well as (Ga,Mn)As films deposited on a GaAs substrate. The ab-initio calculations have been performed within the framework of the local spin density approximation (LSDA) of density functional theory (DFT) using the fully relativistic Korringa-Kohn-Rostoker (KKR) multiple scattering band structure method [19, 20]. For the treatment of the chemical disorder in (Ga,Mn)As alloys we applied the coherent potential approximation (CPA). Moreover, for the bulk and surface calculations we used a regular \vec{k} -mesh of $63 \times 63 \times 63$ points in the full 3D Brillouin Zone (BZ) and 63×63 points in the full 2D BZ, respectively. For the angular momentum expansion of the Green's function a cutoff of $\ell_{\max} = 3$ was applied.

The study of the magneto-crystalline anisotropy (MCA) was performed by calculating the magnetic torque $\vec{T}_i^{(\hat{e}_i)} = -\partial E(\{\hat{e}_k\})/\partial \hat{e}_i \times \hat{e}_i$ acting on the magnetic moment \vec{m}_i of the atomic site i , with a unit vector $\hat{e}_i = \vec{m}_i/|\vec{m}_i|$ pointing along the direction of the magnetization \vec{M} . The component of the magnetic torque with respect to the axis \hat{u}

$$T_{\hat{u}}(\theta, \phi) = -\partial E(\vec{M}(\theta, \phi))/\partial \theta \quad (1)$$

was calculated from first-principles as described in [21]. Here, the \hat{u} vector specified by the angles θ and ϕ (see Fig. 1a) lies within the surface plane and is perpendicular to the direction of the magnetic moment \hat{e}_M . For an uniaxial anisotropy a special geometry can be used which gives a simple relationship between the magnetic torque

and the energy difference between the in-plane and out-of-plane magnetization directions. Setting $\theta = \pi/4$, the torque component $T_{\hat{u}}$ gives the ϕ dependent energy difference $T_{\hat{u}}(\theta = \pi/4, \phi) = E_{||}(\phi) - E_{\perp}$. In the case of an in-plane anisotropy these values can also be used to evaluate the anisotropy energy within the plane, comparing in particular the directions $[110]$ and $[1\bar{1}0]$.

The exchange coupling tensor \underline{J}_{ij} used below for the discussions of the magnetic anisotropy in terms of the anisotropic Mn-Mn exchange interactions [2–4] was calculated as described in Ref. [22]. Here, the effective coefficients of the uniaxial MCA are represented by the following expression [23, 24]:

$$\tilde{K}_i = - \sum_j (J_{ij}^{zz} - J_{ij}^{xx}) + 2K_i, \quad (2)$$

with K_i being the on-site MCA coefficients [23].

In order to study the strain-induced effect in the MCA of deposited (Ga,Mn)As films, we consider at first a bulk system with tetragonal distortion (avoiding surface and interface contributions) which is then characterized by the c/a ratio. Magnetic torque calculations simulating the strain-induced effects in the alloy with 5% Mn yield a linear variation of the magnetic anisotropy energy, $E_{[100]} - E_{[001]}$, from +3.38 to $-3.37 \mu\text{eV}$ per unit cell for c/a ratio varying from 0.99 to 1.01, i.e. the magnetic easy axis changes from an out-of-plane to an in-plane orientation, which is in line with corresponding experimental data [25]. As the $[100]$ and $[010]$ directions are equivalent, this leads to the bi-axial in-plane MCA with $[100]$ and $[010]$ being easy magnetization directions and an in-plane anisotropy energy $E_{[100]} - E_{[110]} \approx -0.1 \mu\text{eV}$ per unit cell.

For a more detailed analysis of the relationship between the MCA and anisotropy of Mn-Mn exchange interactions, calculations of the exchange coupling tensor elements $J_{ij}^{\alpha\beta}$ have been performed for (Ga,Mn)As with 5% Mn both without any distortion as well as with a tetragonal distortion of $c/a = 1.01$. For an undistorted (Ga,Mn)As system we find that the sum $\sum_j (J_{ij}^{\alpha\alpha} - J_{ij}^{\beta\beta})$ ($\alpha, \beta = x, y, z$) over all lattice sites in the expression given in Eq. (2) vanishes (see Fig. 1b). This is a consequence of the system's symmetry, in spite of the fact that the individual terms $(J_{ij}^{\alpha\alpha} - J_{ij}^{\beta\beta})$ with $\alpha \neq \beta$ are non-zero. In the presence of a tetragonal distortion along the z -axis, the symmetry properties within the xy plane (i.e. (001) plane) do not change. Therefore, summation over all lattice sites up to $R_{ij} = 5a$ (with lattice parameter a) shown in Fig. 1c gives $\sum_j (J_{ij}^{xx} - J_{ij}^{yy}) = 0$. For more details, Fig. 1d shows the differences $J_{ij}^{xx} - J_{ij}^{yy}$ for \vec{R}_{ij} taken along $[100]$ and $[010]$ directions (dashed lines). These values are finite and equal in magnitude, but they have an opposite sign and therefore cancel each other upon summation over all sites. However, due to the tetragonal distortion along z , $(J_{ij}^{zz} - J_{ij}^{yy})$ for \vec{R}_{ij} taken along $[001]$

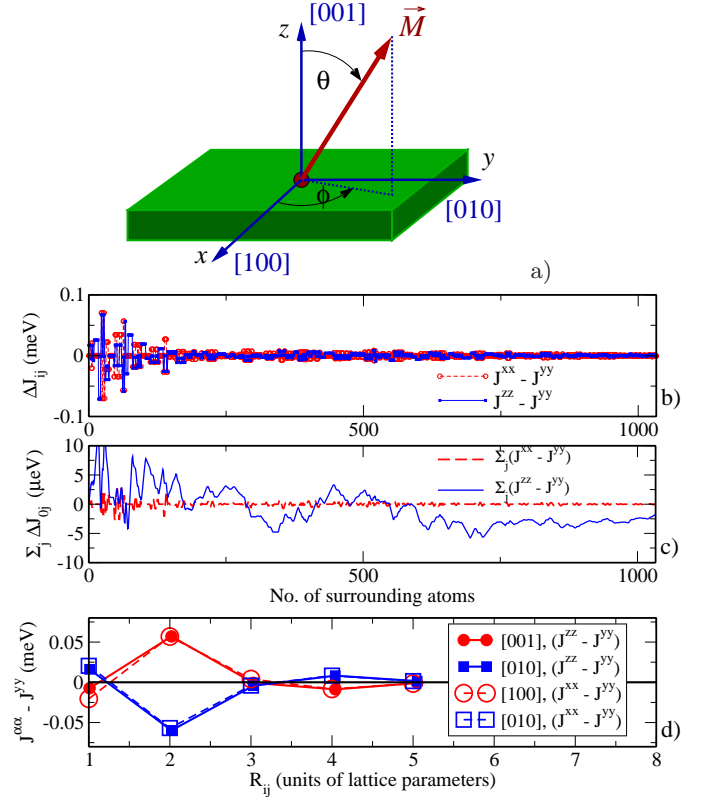


FIG. 1. a) Geometry for the torque calculations; b) $J^{zz} - J^{yy}$ and $J^{xx} - J^{yy}$ for bulk (Ga,Mn)As with 5% Mn, with tetragonal distortion $c/a = 1.01$; c) $\sum_j (J_{ij}^{zz} - J_{ij}^{yy})$ and $\sum_j (J_{ij}^{xx} - J_{ij}^{yy})$ over all lattice sites up to $R_{ij} \leq 5a$; d) $J^{xx} - J^{yy}$ along $[100]$ and $[010]$ directions.

and $[010]$ directions are not equivalent (see Fig. 1d, solid lines) and thus the sum $\sum_j (J_{ij}^{zz} - J_{ij}^{yy})$ over all lattice sites does not vanish anymore. The summation over all lattice sites up to $R_{ij} \leq 5a$ is shown in Fig. 1c which gives the contribution to the uniaxial MCA that originates from the exchange anisotropy being $\approx 2.5 \mu\text{eV}$. Because of the slow convergence of the sum with increasing distance, this gives only an approximation to the true contribution due to the exchange anisotropy. Nevertheless, the value obtained in this way has the same order of magnitude as the MAE obtained from our torque calculations leading to the conclusion that the exchange anisotropy has indeed a significant impact on the total MAE.

Our present investigations of the in-plane uniaxial anisotropy have been performed for a 8 monolayer (ML) thick (Ga,Mn)As film deposited on a semi-infinite (001) -oriented GaAs substrate. In order to distinguish the anisotropy behaviour in the vicinity to the surface with GaAs as well as in the area adjacent to the surface we performed calculations for an uncovered (Ga,Mn)As film as well as one with two additional capping layers of Au. Due to small amount of free charge carriers in (Ga,Mn)As the surface potential decays slowly into bulk leading to

a potential and a charge density gradient within an extended region adjacent to the surface. The existence of such a potential gradient results in the breaking of the 4-fold symmetry of the bulk (Ga,Mn)As system, making the $[1\bar{1}0]$ and $[110]$ directions inequivalent (for the geometry used here this corresponds to the x and y directions, respectively) and leading effectively to a C_{2v} symmetry not only within the few surface/interface layers but also in a rather extended subsurface regime.

We discuss now the surface induced MCA in the film. Here we focus mainly on the MAE properties of a (Ga,Mn)As film with a clean Ga terminated surface deposited on GaAs(001). The results for the energy differences between different magnetization directions are $E_{[110]} - E_{[001]} = -80.56 \mu\text{eV}$ and $E_{[1\bar{1}0]} - E_{[001]} = -32.96 \mu\text{eV}$ per film unit cell (8 ML). This gives an uniaxial in-plane anisotropy with the energy difference of $E_{[110]} - E_{[1\bar{1}0]} = -47.6 \mu\text{eV}$ per film unit cell.

Fig. 2a presents the layer resolved contributions to the $E_{[110]} - E_{[001]}$ and $E_{[1\bar{1}0]} - E_{[001]}$ values, indicated by open and filled symbols, respectively. The difference between these values characterizes the MCA within the plane. One should emphasize here that the contribution to the MCA from the region close to the surface decays slowly into the bulk. Therefore the surface-induced anisotropy effect in the uniaxial in-plane MCA is determined by a rather extended region adjacent to the surface and not just by two or three subsurface layers as it is often observed in metallic systems (e.g. [26]). The corresponding contribution to the energy of the uniaxial in-plane anisotropy exceeds by far the energy of the bi-axial in-plane anisotropy when normalized to the same volume ($E_{[100]} - E_{[110]} \approx -0.1 \mu\text{eV}$ per unit cell of the bulk system). Using these results the MCA of experimental (Ga,Mn)As films consisting of $n + 8$ monolayers can be modelled by combining the contribution of n bulk-like layers with the contribution of 8 layers of surface region. This gives two competing contributions to the MCA: a bi-axial in-plane anisotropy from bulk-like layers of (Ga,Mn)As with a tetragonal distortion and a uniaxial in-plane anisotropy from the area adjacent to the surface. Applying our obtained MAE values to a unit volume, one can get the MCA of the whole film including the surface region. Within our consideration, the coefficient of the in-plane bi-axial anisotropy K_4 does not depend on the film thickness L , while the coefficient of the in-plane uni-axial anisotropy K_2^{\parallel} recalculated per unit volume should decrease with film thickness as $1/L$. Thus, according to our numerical results, a rather strong uniaxial anisotropy should be observed in the case of very thin films, while the increase of the film thickness should lead to a competition of bi-axial and uniaxial anisotropies beginning with a certain film thickness. The contribution from the 'surface' region to the out-of-plane uniaxial anisotropy $E_{[1\bar{1}0]} - E_{[001]}$ decreases as well

with the film thickness as $1/L$. This results in a leading role of the in-plane anisotropy contribution caused by the tetragonal lattice distortion discussed above. Note that an increase of the Mn concentration results in an increase of the charge carriers in the film which again results in better screening of the surface potential. This can be seen in Fig. 2a, where the values $E_{[110]} - E_{[001]}$ and $E_{[1\bar{1}0]} - E_{[001]}$ are shown as a function of the distance from the surface for a (Ga,Mn)As film with 11% Mn. This increase in Mn concentration results in an in-plane MAE $E_{[110]} - E_{[1\bar{1}0]} = -20.8 \mu\text{eV}$ per film unit cell, i.e. one obtains a smaller anisotropy energy when compared to the case of 5%Mn.

Since the uniaxial MCA has its origin in an extended subsurface region one can expect that it is an intrinsic property of the systems and should be observed not only in the case of a clean surface but also in the presence of overlayers on the top of the (Ga,Mn)As film. Corresponding investigations have been performed for a (Ga,Mn)As film with 3 capping layers of Au on top of the (Ga,Mn)As film. The resulting layer resolved contribution to the MCA is shown in Fig. 2b. In spite of the differences in the MAE between the Au capped (Ga,Mn)As film and the case of uncovered film, the general trend in both cases is the same, i.e. one can clearly see that the difference in layer contributions to the MCE for different directions of magnetization, along $[110]$ and $[1\bar{1}0]$, decays slowly with the distance from the surface or Au/(Ga,Mn)As interface, respectively.

To investigate also the effect caused by a concentration gradient along the surface normal within an uncovered (Ga,Mn)As film we dealt with a corresponding film where the Mn concentration varies from 5% at the (Ga,Mn)As/GaAs interface to 6.6% in the surface layer. As can be seen in Fig. 2a such a gradient does not result in a noteworthy change in the MCA.

To analyze in more detail the origin of the surface-induced in-plane uniaxial anisotropy the contribution of the exchange interaction anisotropy in the (Ga,Mn)As film was determined. Fig. 3 shows the difference $J_{ij}^{xx} - J_{ij}^{yy}$ calculated along the $[110]$ and $[1\bar{1}0]$ directions within the film layers where the x and y axes are chosen along $[1\bar{1}0]$ and $[110]$ directions, respectively. As discussed above, for bulk (Ga,Mn)As the variation of $J_{ij}^{xx} - J_{ij}^{yy}$ with distance $|\vec{R}_{ij}|$ is the same for \vec{R}_{ij} along the $[110]$ and $[1\bar{1}0]$ however with different sign. This behaviour is more or less the same for Mn atoms next to the (Ga,Mn)As/GaAs interface (see Fig. 3a). For Mn in the fourth layer (choosing the surface layer as the first layer), however, the situation is changed indicating a pronounced modification of the anisotropic exchange coupling due to the broken symmetry. As a result, the sum over lattice sites in Eq. (2) does not vanish which leads to a contribution to the in-plane uniaxial anisotropy. Fig. 3b shows the corresponding results obtained by summing the terms $(J_{ij}^{xx} - J_{ij}^{yy})$

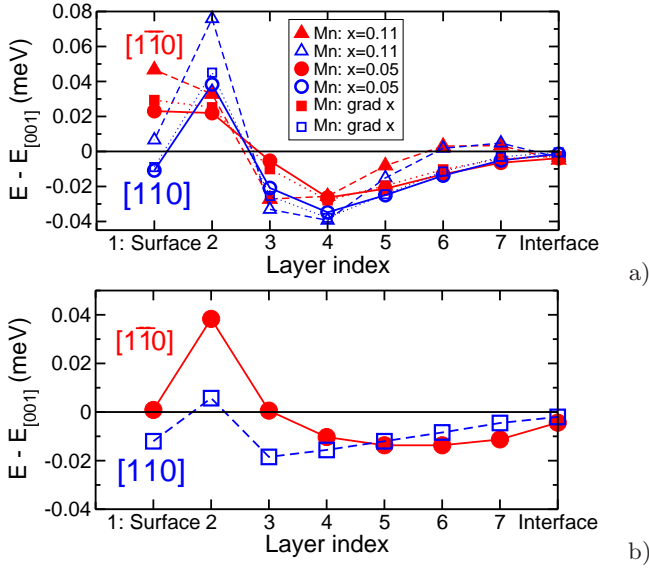


FIG. 2. a) Layer resolved contributions to the MCA energy in the uncovered 8ML (Ga,Mn)As film with 5 at.% Mn (circles) and 11 (triangles) at.% Mn, as well as with Mn content varying from 5 at.% at the interface to 6.6 at.% in surface layer (*grad x*, squares), for two directions of magnetization: $\vec{M}||[110]$ and $\vec{M}||[\bar{1}\bar{1}0]$; b) layer resolved contributions to the MCA energy in the 8ML (Ga,Mn)As film with 5 at.% Mn, with 3 capping layers of Au.

over all lattice sites j within a sphere of radius $2.9a$ with i taken within the layers 1 – 8 in the (Ga,Mn)As film. As one can see, the anisotropy of the exchange interaction gives indeed a substantial contribution to the anisotropy energy $E_{[110]} - E_{[\bar{1}\bar{1}0]}$ for the layers 3 to 8. For the first two film layers, i.e. surface and subsurface layers the two curves strongly deviate reflecting the dominating on-site contribution to the MCA [23, 24].

In summary our results show that the tetragonal distortion (caused by a compressive strain due to lattice mismatch of (Ga,Mn)As and GaAs lattices) is responsible for the bi-axial in-plane anisotropy that is in line with the interpretation given in previous investigations. A strong uniaxial in-plane MCA was found in (Ga,Mn)As film in the area adjacent to the surface or an interface. We conclude that this is a result of the slow decay of the surface potential gradient due to the small amount of free charge carriers. The contribution to the uniaxial in-plane anisotropy decays rather slowly into the bulk and is not restricted to only a few surface layers. Moreover, a significant contribution responsible for the MCA in the films is caused by the anisotropic Mn-Mn exchange interactions mediated by holes in the valence band of (Ga,Mn)As.

Acknowledgements

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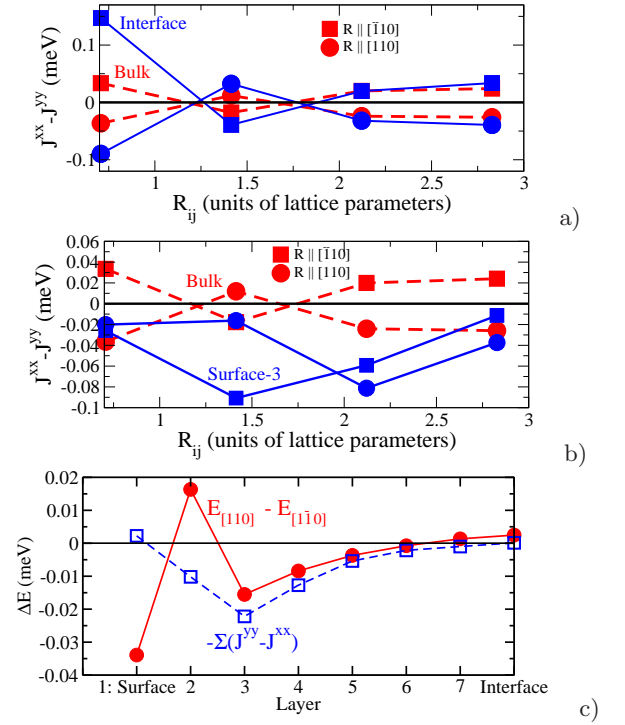


FIG. 3. Variation of $(J_{ij}^{xx} - J_{ij}^{yy})$ with distance R_{ij} of pairs (i, j) of Mn atoms taken in $[110]$ and $[\bar{1}\bar{1}0]$ directions in the (Ga,Mn)As/GaAs film: a) bulk vs (Ga,Mn)As/GaAs interface and b) bulk vs (Surf.- 3)-layer; c) Layer-resolved sum $-\sum_j (J_{ij}^{yy} - J_{ij}^{xx})$ calculated within the sphere of radius $2.9a$ in comparison with the MCA energy $E_{[110]} - E_{[\bar{1}\bar{1}0]}$ evaluated by magnetic torque calculations for the 8ML (Ga,Mn)As film with 5 at.% Mn.

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